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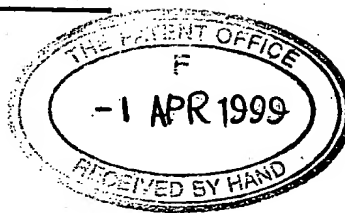
Signed

*AmBrewster*

Dated 6 April 2000

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# Request for grant of a patent

(See the notes on the back of this form. You can also get an explanatory leaflet from the Patent Office to help you fill in this form)

1. Your reference

PPD 50391/GB/P

2. Patent application number

(The P)

9907669.7

- 1 APR 1999

3. Full name and postcode of the or of each applicant (underline all surnames)

ZENECA Limited  
15 Stanhope Gate  
London W1Y 6LN  
UNITED KINGDOM

Patents ADP number (if you know it)

6254007002

If the applicant is a corporate body, give the country/state of its incorporation

UNITED KINGDOM

4. Title of the invention

AGROCHEMICAL COMPOSITION

5. Name of your agent (if you have one)

"Address for service" in the United Kingdom to which all correspondence should be sent (including the postcode)

Michael James RICKS  
Intellectual Property Department  
ZENECA Agrochemicals  
Jealott's Hill Research Station  
P O Box 3538  
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UNITED KINGDOM

Patents ADP number (if you know it)

01282433003

6. If you are declaring priority from one or more earlier patent applications, give the country and the date of filing of the or of each of these earlier applications and (if you know it) the or each application number

Country

Priority application number  
(if you know it)

Date of filing  
(day / month / year)

7. If this application is divided or otherwise derived from an earlier UK application, give the number and the filing date of the earlier application

Number of earlier application

Date of filing  
(day / month / year)

8. Is a statement of inventorship and of right to grant of a patent required in support of this request? (Answer 'Yes' if:

- a) any applicant named in part 3 is not an inventor, or
- b) there is an inventor who is not named as an applicant, or
- c) any named applicant is a corporate body.

See note (d))

Patents Form 1/77

9. Enter the number of sheets for any of the following items you are filing with this form. Do not count copies of the same document

Continuation sheets of this form

Description **7**

Claim(s) **2**

Abstract **1**

Drawing(s) **1**

10. If you are also filing any of the following, state how many against each item.

Priority documents

Translations of priority documents

Statement of inventorship and right to grant of a patent (Patents Form 7/77)

Request for preliminary examination and search (Patents Form 9/77)

Request for substantive examination (Patents Form 10/77)

Any other documents (please specify)

11.

I/we request the grant of a patent on the basis of this application.

**ZENECA LIMITED**

Signature *D. L. Royal*  
Authorised Signatory

Date **01 APRIL 1999**

12. Name and daytime telephone number of person to contact in the United Kingdom

**VIJAYA KUMARI MALLIPEDDI**  
**DAWN LISA ROYAL**

**01344 414365**  
**01344 414079**

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**Notes**

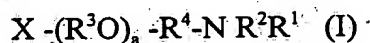
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- Write your answers in capital letters using black ink or you may type them.
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# AGROCHEMICAL COMPOSITION

This invention relates to an agrochemical composition and in particular to an agrochemical composition containing an activity-enhancing adjuvant.

5 An agrochemical is generally used with an adjuvant or combination of adjuvants to provide optimum biological activity. Much has been published on the selection of adjuvants to achieve particular effects with individual agrochemicals and classes of agrochemical. In general it has been assumed that activity-enhancement results from surfactant properties of the adjuvant and most such activity-enhancing adjuvants are surfactants in that they contain  
10 within the molecule both a hydrophobic portion and a lypophobic portion. We have now found that a class of alkoxyated amines provides excellent activity enhancement when used in an agrochemical formulation. Surprisingly certain members of the class have either no surfactant properties or low surfactant properties Furthermore, unlike many surfactants used as conventional agrochemical adjuvants, certain of the alkoxyated amines of the present  
15 invention exhibit an exceptionally low toxicological profile and provide compositions which are especially benign to the environment.

According to the present invention there is provided an aqueous agrochemical composition comprising an agrochemical active ingredient and an adjuvant of formula (I) and salts thereof.



wherein  $R^1$  and  $R^2$  and independently hydrogen or a lower alkyl group or a group  $X-(R^3O)_a-R^4-$  as hereinafter defined wherein X is -OH or a lower alkoxy group containing  
25 from 1 to 6 carbon atoms,

$R^3O$  is an ethoxy, propoxy or butoxy group or a mixture thereof,

$R^4$  is a linear or branched chain alkylene bridging group containing from 1 to 4 carbon atoms X is

(IA) -OH or a lower alkyloxy group containing from 1 to 6 carbon atoms

or X is

(IB) a group  $R^5R^6N-$  or  $R^5R^6N-R^7-$

wherein  $R^5$  and  $R^6$  are independently hydrogen or a lower alkyl group or a group  
or a group  $X-(R^3O)_a-R^4$  as hereinbefore defined wherein X is -OH or a lower alkoxy group  
containing from 1 to 6 carbon atoms,  
and  $R^7$  is a linear or branched chain alkylene bridging group containing from 1 to 4 carbon  
5 atoms

or X is

(IC) a group

$CR^{16}R^{17} - O -$

10 |

$CR^{18} - O - (R^8O)_b - R^9 - NR^{10}R^{11}$

|

$CR^{19}R^{20} - O - (R^{12}O)_c - R^{13} - NR^{14}R^{15}$

wherein  $R^{10}$ ,  $R^{11}$ ,  $R^{14}$  and  $R^{15}$  are independently hydrogen or a lower alkyl group or a group  
15  $X-(R^3O)_a-R^4$  as hereinbefore defined wherein X is -OH or a lower alkoxy group containing  
from 1 to 6 carbon atoms,

and  $R^8$  and  $R^{12}$  are independently alkyl groups containing from 1 to 4 carbon atoms and  $R^9$   
and  $R^{13}$  are independently a linear or branched chain alkylene bridging group containing  
from 1 to 6 carbon atoms,  $R^{16}$ ,  $R^{17}$ ,  $R^{18}$ ,  $R^{19}$  and  $R^{20}$  are independently hydrogen or lower alkyl  
20 groups such as methyl or ethyl or propyl.

and wherein a, is from 1 to 400 or if X is of formula IC, the sum of a, b and c is from 3 to  
400.

A considerable number of salts of the compound of formula (I) suitable for use in  
agrochemical applications will occur one skilled in the art. Examples of salts include  
25 phosphate, sulphate, carboxylate, acetate, formate, chloride, although many other suitable  
salts will occur to one skilled in the art. Alternatively the compound of formula (I) may form  
a salt with an acidic agrochemical such as glyphosate. Salts of the compound of formula (I)  
can also be prepared from acidic surfactants, for example optionally ethoxylated alkyl or  
alkylene ester derivatives of phosphoric acid, or optionally ethoxylated alkyl or alkylene  
30 carboxylic acids or sulphonic acids.

As used herein, the term lower alkyl means a linear or branched chain primary or secondary alkyl group containing from 1 to 6 carbon atoms. Preferred lower alkyl groups contain from 1 to 4 carbon atoms, and methyl, ethyl and propyl or isopropyl groups are especially preferred.

5  $R^1, R^2, R^3, R^6, R^{10}, R^{11}, R^{14}$  and  $R^{15}$  are preferably methyl or hydrogen. Hydrogen is especially preferred.

The compounds for use in the composition of the present invention are alkoxyated monoamines (X is of Formula IA), diamines (X is of Formula IB) or triamines (X is of Formula IC). Alkoxylation typically takes place to introduce ethoxy groups or propoxy groups, although butoxy groups may sometimes also used. Mixed alkoxylation may also take place to introduce for example both ethoxy and propoxy or butoxy groups. Thus each of 10  $-(OR^3)_a$ ,  $-(OR^8)_b$ ,  $-(OR^{12})_c$  and  $-(OR^{21})_d$  in the above formulae independently represent alkoxy groups, for example ethoxy and propoxy groups or a mixture thereof. Thus the definition of group  $-(OR^3)_a$  for example includes a group



wherein a is represented by the sum of x and y. Other combinations of for example ethoxy propoxy and butoxy groups in any desired order are similarly included in the definition of  $-(OR^3)_a$ ,  $-(OR^8)_b$ ,  $-(OR^{12})_c$  and  $-(OR^{21})_d$  respectively

The process of alkoxylation may produce a mixture of products having different 20 degrees of alkoxylation. Thus the values of a, b, and c represent an average degree of alkoxylation over the product as a whole. Preferred values of a, b, and c, respectively are from 1 to 100, for example from 1 to 50.

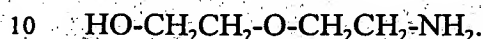
An especially preferred alkoxyated monoamine (wherein X is of formula IA) for use in the present invention has the formula II wherein X is methoxy,  $-(R^3O)_a$  represents a mixture of ethoxy and propoxy groups  $R^4$  represents an propylene bridging group:- 25



The average degree of ethoxylation (x) may vary from 0 to about 45 or more preferably from about 1 to about 40 and the average degree of propoxylation may vary from 0 to about 90 and more preferably from about 1 to about 35. Products are commercially available wherein 30 x is about 1 and y is about 9; x is about 19 and y is about 3; x is about 6 and y is about 29 and x is about 32 and y is about 10. As specific examples of commercially available

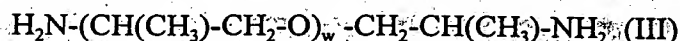
products there may be mentioned JEFFAMINE M600 (JEFFAMINE is a trade mark of Huntsman Limited) having an approximate molecular weight of 600 and a propoxy to ethoxy ratio of 9 to 1, JEFFAMINE M1000 having an approximate molecular weight of 1000 and a propoxy to ethoxy ratio of 3 to 19, JEFFAMINE M2005 having an approximate molecular weight of 2000 and a propoxy to ethoxy ratio of 29 to 6 and JEFFAMINE M2070 having an approximate molecular weight of 2000 and a propoxy to ethoxy ratio of 10 to 32.

Also included in the scope of the present invention is diethylene glycolamine wherein in Formula I, X is OH, (R<sup>3</sup>O) is ethoxy, a is 1 and R<sup>4</sup> is an ethylene bridging group and R<sup>1</sup> and R<sup>2</sup> are hydrogen:-



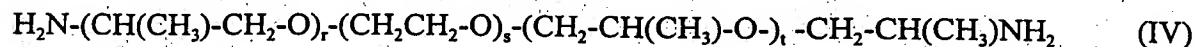
Also included in the scope of the present invention is tris 2-(2-methoxyethoxyl)ethylamine wherein in formula I, X is OCH<sub>3</sub>, (R<sup>3</sup>O) is ethoxy, a is 1 and R<sup>4</sup> is an ethylene bridging group and wherein R<sup>1</sup> and R<sup>2</sup> are each a group -R<sup>4</sup>-(OR<sup>3</sup>)<sub>a</sub>-X wherein (R<sup>3</sup>O) is ethoxy, a is 1, R<sup>4</sup> is an ethylene bridging group and X is OCH<sub>3</sub>.

15 An especially preferred propoxylated diamine (wherein X is of formula IB) for use in the present invention has the formula III wherein X is a group H<sub>2</sub>N-, -(R<sup>3</sup>O-) a represents propoxy groups R<sup>4</sup> represents a propoxy bridging group:-



wherein w is an average of from about 1 to about 80. Products are commercially available wherein w is an average of about 2.6, 5.6, 33.1 and 68. As Examples of commercially available products of formula III, there may be mentioned JEFFAMINE D230 having an approximate molecular weight of 230 and a value of w of about 2.6, JEFFAMINE D400 having an approximate molecular weight of 400 and a value of w of about 5.6, JEFFAMINE D2000 having an approximate molecular weight of 2000 and a value of w of about 33.1, and JEFFAMINE D4000 having an approximate molecular weight of 4000 and a value of w of about 68.

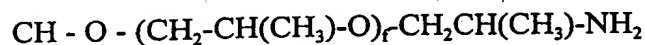
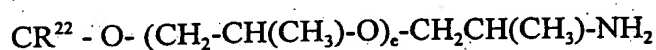
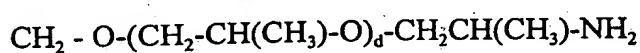
30 An alternative diamine (wherein X is of formula IB) having a mixture of ethoxylation and propoxylation for use in the present invention has the formula IV wherein X is a group H<sub>2</sub>N-, -(R<sup>3</sup>O-) a represents a mixture of ethoxy and propoxy groups R<sup>1</sup> and R<sup>2</sup> are hydrogen and R<sup>4</sup> represents a propylene bridging group:-





wherein the sum of  $r + t$  (total propoxy content) is an average of from 1 to 10 propoxy groups and  $s$  is an average of from 5 to 50 ethoxy units. Commercial products are available wherein the sum of  $r + t$  is about 3.6 and  $s$  represents an average of about 9, or 15.5 respectively or wherein  $r + t$  is about 6 and  $s$  represents an average of about 38.7. As examples of commercially available products of formula (IV) there may be mentioned JEFFAMINE ED 600 having an approximate molecular weight of 600 and an propoxy to ethoxy ratio of 3.6 to 9, JEFFAMINE ED 900 having an approximate molecular weight of 900 and an propoxy to ethoxy ratio of 3.6 to 15.5 and JEFFAMINE ED 2003 having an approximate molecular weight of 2000 and an propoxy to ethoxy ratio of 6.0 to 38.7.

An especially preferred propoxylated triamine (wherein  $X$  is formula IC) has the formula (V)



wherein the sum of  $d + e + f$  is an average of from about 5 to 90 and  $R^{22}$  is hydrogen or lower alkyl such as methyl or ethyl. Products are available commercially wherein the sum of  $d + e + f$  is about 5.6, about 50 and about 85 respectively. Thus as examples of commercially available products of formula (V) there may be mentioned JEFAMINE T403 having an approximate molecular weight of 440 and a value of  $d + e + f$  (total propoxy content) of 5.6, JEFAMINE T3000 having an approximate molecular weight of 3000 and a value of  $d + e + f$  (total propoxy content) of 50 and JEFAMINE T5000 having an approximate molecular weight of 5000 and a value of  $d + e + f$  (total propoxy content) of 85.

The agrochemical for use in the composition of the present invention is preferably an electrolyte or agrochemical salt such as glyphosate or paraquat. Typical of the commonly available salts of glyphosate are the isopropylamine, trimethylsulphonium, sodium, potassium, ammonium, and ethanolamine salts. Paraquat is generally sold in the form of paraquat dichloride.

The proportion of adjuvant of formula (I) to agrochemical, for example glyphosate (expressed as the acid) may vary within wide ranges depending on the desired level of

activation. Typically the proportion of adjuvant of formula (I) to agrochemical will be from 1:20 to 3:1 by weight, for example from 1:10 to 1:1 by weight.

The adjuvant of formula (I) or a mixture of different adjuvants of formula (I) may be used as the sole adjuvant to enhance the biological activity or physical properties of the agrochemical or may alternatively be used in conjunction with one or more additional adjuvants. The adjuvant of formula (I) may for example be combined with cationic surfactants, anionic surfactants, amphoteric surfactants or non-ionic surfactants. Such surfactants are well known in the art but as surfactants which are particularly well suited to be combined with an adjuvant of formula (I) there may be mentioned alkyl glycosides (mono and poly), alcohol ethoxylates, alkyl phenol ethoxylates, alkyl ester ethoxylates, sorbitan ester ethoxylates, siloxane ethoxylates, optionally alkoxyated tertiary or quaternary alkyl amines, alkyl amine oxides, alkyl betaines and sucrose alkyl esters. The adjuvant of formula (I) may also be combined with other activity-enhancing adjuvants, for example ammonium sulphate, urea or humectants, such as glycerol, polyethylene glycol, sorbitol, ethylene glycol, propylene glycol and lactate salts.

The invention is illustrated by the following Examples in which all parts and percentages are by weight unless otherwise stated.

#### Examples 1 to 10

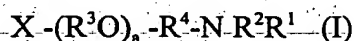
Potassium glyphosate was applied to Abutilon at 500g glyphosate acid equivalent/ha using a spray application volume of 200l/ha in deionised water. The JEFFAMINE adjuvants were used in a proportion 0.2% by weight. A visual assessment of % control was carried out 16 days after treatment.

Example No.	Treatment	% control
Comparison	No adjuvant	37
1	Jeffamine D400	93
2	Jeffamine D2000	76
3	Jeffamine T3000	79
4	Jeffamine ED600	89
5	Jeffamine ED900	85
6	Jeffamine ED2003	85

7	Jeffamine M600	85
8	Jeffamine M1000	81
9	Jeffamine M2005	70
10	Jeffamine M2070	74

## Claims

1. According to the present invention there is provided an aqueous agrochemical composition comprising an agrochemical active ingredient and an adjuvant of formula (I) and salts thereof.



wherein  $R^1$  and  $R^2$  are independently hydrogen or a lower alkyl group or a group  $X-(R^3O)_a-R^4$  as hereinafter defined wherein X is -OH or a lower alkoxy group containing from 1 to 6 carbon atoms

$R^3O$  is an ethoxy, propoxy or butoxy group or a mixture thereof,

$R^4$  is a linear or branched chain alkylene bridging group containing from 1 to 4 carbon atoms X is

(IA) -OH or a lower alkoxy group containing from 1 to 6 carbon atoms or X is

(IB) a group  $R^5R^6N$  or  $R^5R^6N-R^7$

wherein  $R^5$  and  $R^6$  are independently hydrogen or a lower alkyl group or a group or a group  $X-(R^3O)_a-R^4$  as hereinbefore defined wherein X is -OH or a lower alkoxy group containing from 1 to 6 carbon atoms,

and  $R^7$  is a linear or branched chain alkylene bridging group containing from 1 to 4 carbon atoms.

or X is

(IC) a group

$CR^{16}R^{17}-O-$

|

$CR^{18}-O-(R^8O)_b-R^9-NR^{10}R^{11}$

|

$CR^{19}R^{20}-O-(R^{12}O)_c-R^{13}-NR^{14}R^{15}$

wherein  $R^{10}$ ,  $R^{11}$ ,  $R^{14}$  and  $R^{15}$  are independently hydrogen or a lower alkyl group or a group  $X-(R^3O)_a-R^4$  as hereinbefore defined wherein X is -OH or a lower alkoxy group containing from 1 to 6 carbon atoms, and  $R^8$  and  $R^{12}$  are independently alkyl groups containing from 1 to 4 carbon atoms and  $R^9$  and  $R^{13}$  are independently a linear or branched chain alkylene bridging group containing from 1 to 6 carbon atoms,  $R^{16}$ ,  $R^{17}$ ,  $R^{18}$ ,  $R^{19}$  and  $R^{20}$  are independently hydrogen or lower alkyl groups such as methyl or ethyl or propyl and wherein a, is from 1 to 400 or if X is of formula IC, the sum of a, b and c is from 3 to 400.

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